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(54) [Title of the Invention] ORGANIC EL ELEMENT

(57) [Abstract]

[Object] To provide an organic EL element which can be stably driven for a long period.

[Structure] An organic EL element formed by sequentially stacking an anode, an organic light emitting layer, and a cathode is characterized in that an interface layer

made from an aromatic amine compound is provided between the organic light emitting layer and the cathode.

[Effect] Light emission with luminance, which is sufficient in practical use, can be obtained at low driving voltage and a stable light emitting performance is shown for a long period.

[Scope of Claim]

[Claim 1] An organic EL element formed by sequentially stacking an anode, an organic light emitting layer, and a cathode, characterized in that an interface layer made from an aromatic amine compound is provided between the organic light emitting layer and the cathode.

[Detailed Description of the Invention]

[0001]

[Industrial Application Field] The present invention relates to an organic EL element, and in particular, relates to a thin-film device, which emits light when applying electric field to a light emitting layer made from an organic compound.

[0002]

[Conventional Art] As a thin-film electroluminescence (EL) element, an element in which Mn or a rare earth element (such as Eu, Ce, Tb, and Sm), which is a light emitting center, is doped into ZnS, CaS, SrS, or the like, which is II-VI group compound semiconductor of an inorganic material, has been conventionally used in general. However, an EL element manufactured using the above-mentioned inorganic material has the following problems:

(1) alternating current drive is required (50 to 1,000 Hz);

(2) high driving voltage (about 200 V);

(3) full colorization is difficult (in particular, blue is difficult); and

(4) cost of a peripheral driver circuit is high.

[0003] However, in order to solve the above problems, development of an EL element using an organic thin film has been carried out in recent years. In particular, because of development of an organic EL element in which the type of an electrode is optimized to improve carrier injecting efficiency from the electrode to improve light emitting

efficiency and an organic hole transporting layer formed using aromatic diamine and a light emitting layer formed using an aluminum complex of 8-hydroxyquinoline are provided (Appl. Phys. Lett., vol. 51, p.p. 913, 1987), the light emitting efficiency is drastically improved as compared with a conventional EL element formed using a single crystal such as anthracene.

[0004] Further, as an organic light emitting layer, high molecular materials such as poly(p-phenylenevinylene) (Nature, vol. 347, p.p. 539, 1990; Appl. Phys. Lett., vol. 61, p.p. 2,793, 1992); poly[2-methoxy,5-(2'-ethylhexoxy)-1,4-phenylenevinylene] (Appl. Phys. Lett., vol. 58, p.p. 1982, 1991; Thin Solid Films, vol. 216, p.p. 96, 1992; Nature, vol. 357, p.p. 477, 1992); and poly(3-alkylthiophene) (Jpn. J. Appl. Phys., vol. 30, p.p. L1,938, 1991; J. Appl. Phys., vol. 72, p.p. 564, 1992) have been developed. An element in which a light emitting material and an electron transfer material are mixed in a high molecule such as polyvinylcarbazole (Applied Physics, vol. 61, p.p. 1,044, 1992) has also been developed.

[0005] In such an organic EL element as shown above, a transparent electrode such as indium tin oxide (ITO) is generally used as an anode whereas a metal electrode having a low work function is generally used as a cathode so as to inject electrons efficiently, and a magnesium alloy, calcium, or the like is used. A biggest problem of an organic EL element is duration of life. One of factors limiting the duration of life is generation of dark spots (which indicate non-light emitting portions of a light emitting portion of the element) derived from a cathode material. Therefore, in a case of saving an organic EL element for long time, the number and the size of dark spots inside the organic EL element are increased, thereby shortening duration of life of the organic EL element.

[0006]

[Problems to be Solved by the Invention] In an organic EL element, which has been already disclosed, EL emission is generated by recombination of holes injected from an anode and electrons injected from a cathode. Generally, in a case of injecting electrons as carriers, the electrons must be injected such that the electrons overcome an injection barrier in an interface between a cathode and an organic light emitting layer. In order to lower the electron injection barrier so as to improve injection efficiency, a metal

electrode having a low work function such as a magnesium alloy or calcium has been used as a cathode. However, since such a metal material is not well-adhered to an organic light emitting layer, the metal material is separated from the organic light emitting layer, and in forming or after forming the cathode over the organic light emitting layer by evaporation, a reaction is generated between a material of the cathode and a material of the organic light emitting layer. This results in deterioration in a light emitting characteristic or generation of dark spots of an element during a period of keeping the element for long time.

[0007] In view of the above circumstances, the present inventors keenly examined to be intended to provide an organic El element, which can maintain a stable light emitting characteristic for long time and prevent generation of dark spots. As a result of the examination, they found that it is preferable to provide an interface layer formed using an aromatic amine compound between an organic light emitting layer and a cathode, which results in completion of the present invention.

[0008]

[Means for Solving the Problem] That is, in an aspect of the present invention, an organic EL element is formed by sequentially stacking an anode, an organic light emitting layer, and a cathode, and an interface layer formed using an aromatic amine compound is provided between the organic light emitting layer and the cathode. The organic EL element of the present invention will be described below with reference to the attached drawings.

[0009] FIG. 1 is a schematic cross sectional view showing a structural example of an organic EL element of the present invention. Reference numeral 1 represents a substrate, reference numerals 2a and 2b represent conductive layers, reference numeral 3 represents an organic light emitting layer, and reference numeral 4 represents an interface layer. The substrate 1 serves as a supporting body of the organic EL element of the present invention. As the substrate 1, a plate of quartz or glass, a metal plate, a metal foil, a plastic film, a sheet, or the like is used. A transparent synthetic resin substrate such as polyester, polymetaacrylate, polycarbonate, or polysulfone is preferable.

[0010] The conductive layer 2a is provided over the substrate 1. The conductive layer 2a is generally formed using metal such as aluminum, gold, silver, nickel, palladium, or tellurium; metal oxide such as oxide of indium and/or tin; copper iodide; carbon black; or a conductive high molecule such as poly(3-methylthiophene). The conductive layer is generally formed by sputtering, vacuum evaporation, or the like. In a case of using a fine metal particle such as silver, copper iodide, carbon black, a metal oxide fine particle having a conducting property, a fine powder of a conductive high molecule, or the like, the conductive layer can also be formed in such a way that the material is dispersed in an appropriate binder resin solution and the solution is applied to a surface of the substrate. Further, in a case of using a conductive high molecule, the conductive layer can also be formed in such a way that a thin film is directly formed on the substrate by electric field polymerization or by application (Appl. Phys. Lett., vol. 60, p.p. 2,711, 1992). The above conductive layer can also be formed by stacking layers using different substances. The thickness of the conductive layer 2a is differed depending on its light transmitting property to be required. When a light transmitting property is required, transmittance of visible light is desirably 60% or more, and preferably 80% or more. In this case, the thickness is generally set to be 50 to 10,000 Å, and preferably, about 100 to 5,000 Å.

[0011] In a case where the conductive layer 2a may be opaque, it may be formed by the same material as the substrate 1. Further, the above conductive layer can be formed by stacking different substances. In the example of FIG. 1, the conductive layer 2a serves as an anode having a function of injecting holes. On the other hand, the conductive layer 2b serves as a cathode having a function of injecting electrons to the organic light emitting layer 3 through the interface layer 4. As a material used for the conductive layer 2b, the materials for the conductive layer 2a as described above can be used. In order to inject electrons efficiently, metal having a low work function is preferable, and appropriate metal such as tin, magnesium, indium, aluminum, or silver, or an alloy thereof is used. The thickness of the conductive layer 2b is generally the same as that of the conductive layer 2a. Further, though not shown in FIG. 1, a substrate, which is the same as the substrate 1, can be provided over the conductive layer 2b. Note that it is necessary for an EL element that

at least one of the conductive layers 2a and 2b has an excellent light transmitting property. Accordingly, it is desired that one of the conductive layers 2a and 2b preferably has the thickness of 100 to 5,000 Å so as to have an excellent light transmitting property.

[0012] The organic light emitting layer 3 is provided over the conductive layer 2a. The organic light emitting layer 3 is formed by using a material in which between the electrodes applied with electric field, holes injected from the anode and electrons injected from the cathode are efficiently transported to be recombined and light is efficiently emitted by the recombination. In order to improve light emitting efficiency, the organic light emitting layer 3 is generally divided into a hole transporting layer 3a and an electron transporting layer 3b as shown in FIG. 2 so that the organic light emitting layer has divided functions (Appl. Phys. Lett., vol. 51, p.p. 913, 1987).

[0013]

In the above-mentioned functional separation type element, a hole transporting material is necessary to be a material having high hole injecting efficiency from the conductive layer 2a and being capable of efficiently transporting injected holes. Therefore, it is required that the hole transporting material has low ionization potential and high hole mobility along with excellent stability, in which an impurity, which becomes a trap, is not easily generated in manufacturing or in use.

[0014] As such a hole transporting compound, for example, aromatic amine compounds disclosed in the 13^{th} to 14^{th} sections of Japanese Patent Application Laid-Open No. S59-194393 and US Patent No. 4,175,960 such as N,N'diphenyl-N,N'-(3-methylphenyl)-1,1'-biphenyl-4,4'-diamine; 1,1'-bis(4-di-p-tolylaminophenyl)cyclohexane; and 4,4'-bis(diphenylamino)quadrophenyl; a hydrazone compound disclosed in Japanese Patent Application Laid-Open No. H2-311591; a silazane compound disclosed in US Patent No. 4,950,950; and the like can be given. These compounds may be independently used or each of them may be used by mixing with each compound, if required. In addition to the above mentioned compounds, high molecular materials such as polyvinylcarbazole and polysilane (Appl. Phys. Lett., vol. 59, p.p. 2,760, 1991) can be given.

[0015] A hole transporting layer 3a is formed by stacking the above-mentioned organic hole transporting material over the conductive layer 2a by application or vacuum evaporation. In the case of application, an application solution in which an additive such as a binder resin, which does not become a trap of holes, or an application improver, e.g., a leveling agent or the like, is added and dissolved in one or more kinds of organic hole transporting compounds, is controlled. The controlled application solution is applied to the surface of the conductive layer 2a by a method such as spin coating, and then is dried to form the organic hole transporting layer 3. As the binder resin, polycarbonate, polyarylate, polyester, and the like can be given. When the binder resin is added with too much additive, hole mobility is reduced, and therefore, the amount of an additive is desirably low as much as possible and is preferably set to be 50 weight% or less.

[0016] In the case of the vacuum evaporation, an organic hole transporting material is put in a crucible installed in a vacuum container, and the vacuum container is evacuated by an appropriate vacuum pump such that the pressure is reduced to be 10^{-6} Torr. Thereafter, the crucible is heated and the hole transporting material is evaporated to form a layer over a substrate placed to face the crucible. The thickness of the hole transporting layer 3a is generally 100 to 3,000 Å, and preferably, 300 to 1,000 Å. To uniformly form a thin film like this, vacuum evaporation is usually employed.

[0017] As a material of the hole transporting layer 3a, an inorganic material can be used instead of an organic compound. An inorganic material is required to have the same condition as an organic hole transporting compound. As an inorganic material used for the hole transporting layer 3, p-type hydrogenated amorphous silicon, p-type hydrogenated amorphous silicon carbide, p-type hydrogenated microcrystalline silicon carbide, p-type zinc sulfide, p-type zinc selenide, and the like can be given. An inorganic hole transporting layer made from these materials is formed by CVD, plasma CVD, vacuum evaporation, sputtering, or the like.

[0018] The thickness of the inorganic hole transporting layer is generally 100 to 3,000 Å, and preferably, 300 to 1,000 Å as well as the organic hole transporting layer. An electron transporting layer 3b is formed over the hole transporting layer 3a. The electron

transporting layer 3b is formed of a compound, which can efficiently transport electrons toward the hole transporting layer from the cathode between electrodes applied with an electric field.

[0019] As an organic electron transporting compound, a compound, which has high injecting efficiency of electrons from the conductive layer 2b and can efficiently transport the injected electrons, is required. Therefore, such a compound is required to be a compound having high electron affinity and high electron mobility along with excellent stability, in which an impurity, which becomes a trap, is not easily generated in manufacturing or in use.

[0020] As a material satisfying these conditions, an aromatic compound such as tetraphenyl butadiene (Japanese Patent Application Laid-Open No. Sho 57-51781); a metal complex such as an aluminum complex like 8-hydroxyquinoline (Japanese Patent Application Laid-Open No. S59-194393); a cyclopentadiene derivative (Japanese Patent Application Laid-Open No. H2-289675); a perinon derivative (Japanese Patent Application Laid-Open No. H2-289676); an oxadiazole derivative (Japanese Patent Application Laid-Open No. H2-216791); a bis-styrylbenzene derivative (Japanese Patent Application Laid-Open Nos. H1-245087 and H2-222484); a perylene derivative (Japanese Patent Application Laid-Open Nos. H2-189890 and H 3-791); a coumarin compound (Japanese Patent Application Laid-Open Nos. H2-191694 and H3-792); a rare earth complex (Japanese Patent Application Laid-Open No. H1-256584); a distyrylpyrazine derivative (Japanese Patent Application Laid-Open No. H2-252793); a p-phenylene compound (Japanese Patent Application Laid-Open No. H3-33183); a thiadiazolopyridine derivative (Japanese Patent Application Laid-Open No. H3-37292); a pyrrolopyridine derivative (Japanese Patent Application Laid-Open No. H3-37293); a naphthyridine derivative (Japanese Patent Application Laid-Open No. H3-203982); and the like can be given.

[0021] An organic electron transporting layer using such a compound serves to transport electrons and emit light upon recombination of holes and electrons. The organic electron transporting layer also serves as a light emitting layer. When an organic hole transporting compound has a light emitting function, the organic electron transporting layer only serve to transport electrons.

[0022] In order to change color of light emission along with improvement of light emitting efficiency of an element, for example, it is performed that an aluminum complex of 8-hydroxyquinoline which is a host material is doped with a fluorescent dye for a laser such as coumarin (J. Appl. Phys., vol. 65, p.p. 3,610, 1989). In the present invention, when the above-mentioned organic electron transporting material is used as a host material and various kinds of fluorescent dyes are doped in the host material at 10^{-3} to 10 mol%, a light emitting characteristic of an element can be further improved. The thickness of the electron transporting layer 3b is generally 100 to 2,000 Å, and preferably, 300 to 1,000 Å.

[0023] Although the organic electron transporting layer can also be formed by the same method as the organic hole transporting layer, the organic electron transporting layer is generally formed by vacuum evaporation. As a method for further improving light emitting efficiency of an organic EL element, it is considered that another electron transporting layer 3c is further stacked on the electron transporting layer 3b (see FIG. 3). As a compound used for the electron transporting layer 3c, a compound having further strong electron transporting ability, to which electrons from the cathode are easily injected, is required. As such an electron transporting material, the following materials can be given:

[0024]

[Chemical formula 1]

[0025] an oxadiazole derivative such as the chemical formula 1 (Appl. Phys. Lett., vol. 55, p.p. 1,489, 1989); a system in which the oxadiazole derivative is dispersed in a resin such as PMMA (Appl. Phys. Lett., vol. 61, p.p. 2,793, 1992); n-type hydrogenated amorphous silicon carbide; n-type zinc sulfide; n-type zinc selenide; and the like. The thickness of the electron transporting layer 3c is generally 50 to 2,000 Å, and preferably, 100 to 1,000 Å.

[0026] Further, as a material of a single-layer type organic light emitting layer 3 having a single function, there are high molecular materials such as the above-mentioned poly(p-phenylenevinylene) (Nature, vol. 347, p.p. 539, 1990; Appl. Phys. Lett., vol. 61, p.p. 2,793, 1992), poly[2-methoxy, 5-(2'-ethylhexoxy)-1,4-phenylenevinylene] (Appl. Phys. Lett., vol. 58, p.p. 1982, 1991; Thin Solid Films, vol. 216, p.p. 96, 1992; Nature, vol. 357,

p.p. 477, 1992), and poly(3-alkylthiophene) (Jpn. J. Appl. Phys. vol. 30, p.p. L1,938, 1991; J. Appl. Phys. vol. 72, p.p. 564, 1992); and a system in which a light emitting material and an electron transporting material are mixed in a high molecule such as polyvinyl carbazole (Applied Physics, vol. 61, p.p. 1,044, 1992).

[0027] An interface layer 4 is formed over the organic light emitting layer. As roles of the interface layer, it can be given that interface layer 4 have an affinity to the organic light emitting layer, good adhesion to the cathode, chemical stability, and an effect of preventing reaction between the organic light emitting layer and the cathode in and/or after forming the cathode. Further, it is important that the interface layer have an even thin-film shape from the viewpoint of adhesion with the cathode. The present inventors found out that an aromatic amine compound is preferable as a material having such roles.

[0028] The interface layer made from an aromatic amine compound is formed by application or vacuum evaporation as well as the above described organic hole transporting material. In the present invention, as set forth above, by using an aromatic amine compound as a material of the interface layer between the organic light emitting layer and the cathode of the organic EL element, excellent stability of the element can be achieved.

[0029] As preferable aromatic amine compounds, compounds represented by the following general formulas (I) to (V) can be given:

[0030]

[Chemical formula 2]: general formula (I)

[0031] (in the general formula (I), A represents a divalent hydrocarbon residue, which may have a substituent, or a direct binding. Each of R^1, R^2, R^3, and R^4 represents a substituted or unsubstituted alkyl group, a substituted or unsubstituted cycloalkyl group, a substituted or unsubstituted alkenyl group, a substituted or unsubstituted aryl group, or an aralkyl group, and R^1, R^2, R^3, and R^4 may be identical to or different from one another. Each of R^5 and R^6 represents a hydrogen atom, a halogen atom, a hydroxyl group, a substituted or unsubstituted alkyl group, a substituted or unsubstituted cycloalkyl group, a substituted or unsubstituted alkenyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted alkoxy group, a substituted or unsubstituted dialkylamino group, or a substituted or unsubstituted

diarylamino group.)

[0032]

[Chemical formula 3]: general formula (II)

[0033] (in the general formula (II), each of R^7, R^8, R^9, and R^{10} represents a substituted or unsubstituted alkyl group, a substituted or unsubstituted cycloalkyl group, a substituted or unsubstituted alkenyl group, a substituted or unsubstituted aryl group, or an aralkyl group, and R^7, R^8, R^9, and R^{10} may be identical to or different from one another. Each of R^{11} and R^{12} represents a hydrogen atom, a halogen atom, or a hydroxyl group. Alternatively, each of R^{11} and R^{12} represents a saturated or unsaturated aliphatic hydrocarbon group, an aromatic hydrocarbon group, an alkoxy group, an aryloxy group, a dialkylamino group, or a diarylamino group, which may individually have a substituent. X represents a divalent hydrocarbon residue, which may have a substituent.)

[0034]

[Chemical formula 4]: general formula (III)

[0035] (in the general formula (III), each of R^{13}, R^{14}, R^{15}, and R^{16} represents a substituted or unsubstituted alkyl group, a substituted or unsubstituted cycloalkyl group, a substituted or unsubstituted alkenyl group, a substituted or unsubstituted aryl group, or an aralkyl group. R^{13}, R^{14}, R^{15}, and R^{16} may be identical to or different from one another. Each of R^{17} and R^{18} represents a hydrogen atom, a halogen atom, or a hydroxyl group. Alternatively, each of R^{17} and R^{18} represents a saturated or unsaturated aliphatic hydrocarbon group, an aromatic hydrocarbon group, an alkoxy group, an aryloxy group, a dialkylamino group, or a diarylamino group, which may individually have a substituent. Y and Z individually represent a divalent hydrocarbon residue, which may have a substituent, or a direct binding. Y and Z may be identical to or different from each other.)

[0036]

[Chemical formula 5]: general formula (IV)

[0037] (in the general formula (IV), B represents a trivalent hydrocarbon residue, which may have a substituent, or a nitrogen atom. A plurality of R^{19} individually represent a substituted or unsubstituted alkyl group, a substituted or unsubstituted cycloalkyl group, a substituted or unsubstituted alkenyl group, a substituted or unsubstituted aryl group, or an

aralkyl group. The plurality of R^{19} may be identical to or different from one another. A plurality of R^{20} individually represent a hydrogen atom, a halogen atom, a hydroxyl group, a substituted or unsubstituted alkyl group, a substituted or unsubstituted cycloalkyl group, a substituted or unsubstituted alkenyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted alkoxy group, a substituted or unsubstituted dialkylamino group, or a substituted or unsubstituted diarylamino group.)

[0038]

[Chemical formula 6]: general formula (V)

[0039] (in the general formula (V), D represents a trivalent hydrocarbon residue, which may have a substituent, or a nitrogen atom. A plurality of R^{21} individually represent a substituted or unsubstituted alkyl group, a substituted or unsubstituted cycloalkyl group, a substituted or unsubstituted alkenyl group, a substituted or unsubstituted aryl group, or an aralkyl group. The plurality of R^{21} may be identical to or different from one another. A plurality of R^{22} individually represent a hydrogen atom, a halogen atom, a hydroxyl group, a substituted or unsubstituted alkyl group, a substituted or unsubstituted cycloalkyl group, a substituted or unsubstituted alkenyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted alkoxy group, a substituted or unsubstituted dialkylamino group, or a substituted or unsubstituted diarylamino group.) In the general formula (I), A preferably represents an alkylene group, which may have a substituent such as $-CH_2-$, $-CH_2-CH_2-$, or $-CH_2CH_2CH_2-$; a cycloalkylene group such as a cyclohexylene group; an alkenylene group, which may have a substituent such as -CH=CH-, -C(CH3)=CH-, $-CH_2-CH=CH-CH_2-$, or -CH=CH-CH=CH-; an arylene group, which may have a substituent, such as a phenylene group, a naphthylene group, and a phenanthlene group; or a direct binding. Each of R^1, R^2, R^3, and R^4 represents an alkyl group, which may have a substituent, such as a methyl group, an ethyl group, a propyl group, a butyl group, and a hexyl group; a cycloalkyl group such as a cyclohexyl group; an alkenyl group such as a vinyl group; a phenyl group, which may have a substituent, such as an alkyl group, an alkoxy group, a halogen atom, an amino group, and an aryl group; an aryl group such as a naphthyl group; or an aralkyl group such

as a benzyl group, a naphthylmethyl group, and a phenethyl group. R^1, R^2, R^3, and R^4 may be identical to or different from one another. Each of R^5 and R^6 represents an alkyl group, which may have a substituent such as a hydrogen atom, a halogen atom, and a hydroxyl group, such as a methyl group, an ethyl group, a propyl group, a butyl group, and a hexyl group; a cycloalkyl group such as a cyclohexyl group; an alkenyl group such as a vinyl group; a phenyl group, which may have a substituent such as an alkyl group, an alkoxy group, a halogen atom, an amino group, and an aryl group; an aryl group such as a naphthyl group; an aralkyl group such as a benzyl group, a naphthylmethyl group, and a phenethyl group; an alkoxy group such as a methoxy group, an ethoxy group, and a butoxy group; a dialkylamino group such as a dimethylamino group and a diethylamino group; and a diarylamino group such as a dibenzylamino group and a diphenylamino group. R^5 and R^6 may be identical to or different from each other.

[0040] Next, main specific examples of compounds represented by the general formula (I) are shown in Table 1 and Table 2 below; however, compounds represented by the general formula (I) are not limited thereto.

[0041]

[Table 1]

[0042]

[Table 2]

[0043] In the general formula (II), it is preferable that each of R^7, R^8, R^9, and R^{10} represent an alkyl group, which may have a substituent, such as a methyl group, an ethyl group, a propyl group, a butyl group, and a hexyl group; a cycloalkyl group such as a cyclohexyl group; an alkenyl group such as an allyl group; a phenyl group, which may have a substituent, such as an alkyl group, an alkoxy group, a halogen atom, an amino group, and an aryl group; an aryl group such as a naphthyl group; or an aralkyl group such as a benzyl group, a naphthylmethyl group, and a phenethyl group. R^7, R^8, R^9, and R^{10} may be identical to or different from one another. Each of R^{11} and R^{12} represents an alkyl group, which may have a substituent such as a hydrogen atom, a halogen atom, and a hydroxyl group, such as a methyl group, an ethyl group, a propyl group, a butyl group, and a hexyl group; a cycloalkyl group such as a cyclohexyl group; an alkenyl group such as a

vinyl group; a phenyl group, which may have a substituent, such as an alkyl group, an alkoxy group, a halogen atom, an amino group, and an aryl group; an aryl group such as a naphthyl group; an aralkyl group such as a benzyl group, a naphthylmethyl group, and a phenethyl group; an alkoxy group such as a methoxy group, an ethoxy group, and a butoxy group; a dialkylamino group such as a dimethylamino group and a diethylamino group; and a diarylamino group such as a dibenzylamino group and a diphenylamino group. R^{11} and R^{12} may be identical to or different from each other. X represents a divalent hydrocarbon residue such as a methylene group, a propylene group, a xylene group, a cycloxylene group, a vinylene group, a phenylene group, and -CO-, and each of them may have a substituent such as a halogen atom, a hydroxyl group, a saturated or unsaturated hydrocarbon group, an alkoxy group, an aryloxy group, a dialkylamino group, and a diarylamino group.

[0044] Next, main specific examples of compounds represented by the general formula (II) are shown in Table 3 and Table 4 below; however, compounds represented by the general formula (II) are not limited thereto.

[0045]

[Table 3]

* Ortho-position with respect to amino group.

[0046]

[Table 4]

[0047] In the general formula (III), it is preferable that each of R^{13}, R^{14}, R^{15}, and R^{16} represent an alkyl group, which may have a substituent, such as a methyl group, an ethyl group, a propyl group, a butyl group, and a hexyl group; a cycloalkyl group such as a cyclohexyl group; an alkenyl group such as an allyl group; a phenyl group, which may have a substituent such as an alkyl group, an alkoxy group, a halogen atom, an amino group, and an aryl group; an aryl group such as a naphthyl group; or an aralkyl group such as a benzyl group, a naphthylmethyl group, and a phenethyl group. R^{13}, R^{14}, R^{15}, and R^{16} may be identical to or different from one another. Each of R^{17} and R^{18} represents an alkyl group, which may have a substituent such as a hydrogen atom, a halogen atom, and a hydroxyl group, such as a methyl group, an ethyl group, a propyl group, a butyl group, and

a hexyl group; a cycloalkyl group such as a cyclohexyl group; an alkenyl group such as a vinyl group; a phenyl group, which may have a substituent such as an alkyl group, an alkoxy group, a halogen atom, an amino group, and an aryl group; an aryl group such as a naphthyl group; an aralkyl group such as a benzyl group, a naphthylmethyl group, and a phenethyl group; an alkoxy group such as a methoxy group, an ethoxy group, and a butoxy group; a dialkylamino group such as a dimethylamino group and a diethylamino group; or a diarylamino group such as a dibenzylamino group and a diphenylamino group. R^{17} and R^{18} may be identical to or different from each other. Each of Y and Z represents a divalent hydrocarbon residue such as a methylene group, a propylene group, a xylene group, a cycloxylene group, a vinylene group, and a phenylene group; or a direct binding. Each of them may have a substituent such as a halogen atom, a hydroxyl group, a saturated or unsaturated hydrocarbon group, an alkoxy group, an aryloxy group, a dialkylamino group, and a diarylamino group.

[0048] Next, a main specific example of a compound represented by the general formula (III) is shown in Table 5 below; however, a compound represented by the general formula (III) is not limited thereto.

[0049]

[Table 5]

* Ortho-position with respect to amino group.

[0050] In the general formula (IV), it is preferable that B represent -CH=; a benzene residue with the exception of a hydrogen atom in the 1-, 3-, and 5-positions; a cyclohexane residue with the exception of a hydrogen atom in the 1-, 3-, and 5-positions; or a nitrogen atom. The plurality of R^{19} individually represent an alkyl group, which may have a substituent, such as a methyl group, an ethyl group, a propyl group, a butyl group, and a hexyl group; a cycloalkyl group such as a cyclohexyl group; an alkenyl group such as a vinyl group; a phenyl group, which may have a substituent such as an alkyl group, an alkoxy group, a halogen atom, an amino group, and an aryl group; an aryl group such as a naphthyl group; or an aralkyl group such as a benzyl group, a naphthylmethyl group, and a phenethyl group. The plurality of R^{20} individually represent an alkyl group, which may have a substituent such as a hydrogen atom, a halogen atom, and a hydroxyl group, such as

a methyl group, an ethyl group, a propyl group, a butyl group, and a hexyl group; a cycloalkyl group such as a cyclohexyl group; an alkenyl group such as a vinyl group; a phenyl group, which may have a substituent such as an alkyl group, an alkoxy group, a halogen atom, an amino group, and an aryl group; an aryl group such as a naphthyl group; an aralkyl group such as a benzyl group, a naphthylmethyl group, and a phenethyl group; an alkoxy group such as a methoxy group, an ethoxy group, and a butoxy group; a dialkylamino group such as a dimethylamino group and a diethylamino group; or a diarylamino group such as a dibenzylamino group and a diphenylamino group.

[0051] Next, a main specific example of a compound represented by the general formula (IV) is shown in Table 6 below; however, a compound represented by the general formula (IV) is not limited thereto.

[0052]

[Table 6]

* Ortho-position with respect to amino group.

[0053] In the general formula (V), it is preferable that D represent -CH=; a benzene residue with the exception of a hydrogen atom in the 1-, 3-, and 5-positions; a cyclohexane residue with the exception of a hydrogen atom in the 1-, 3-, and 5-positions; or a nitrogen atom. The plurality of R^{21} individually represent an alkyl group, which may have a substituent, such as a methyl group, an ethyl group, a propyl group, a butyl group, and a hexyl group; a cycloalkyl group such as a cyclohexyl group; an alkenyl group such as a vinyl group; a phenyl group, which may have a substituent such as an alkyl group, an alkoxy group, a halogen atom, an amino group, and an aryl group; an aryl group such as a naphthyl group; and an aralkyl group such as a benzyl group, a naphthylmethyl group, and a phenethyl group. The plurality of R^{22} individually represent an alkyl group, which may have a substituent such as a hydrogen atom, a halogen atom, and a hydroxyl group, such as a methyl group, an ethyl group, a propyl group, a butyl group, and a hexyl group; a cycloalkyl group such as a cyclohexyl group; an alkenyl group such as a vinyl group; a phenyl group, which may have a substituent such as an alkyl group, an alkoxy group, a halogen atom, an amino group, and an aryl group; an aryl group such as a naphthyl group; an aralkyl group such as a benzyl group, a naphthylmethyl group, and a phenethyl group;

an alkoxy group such as a methoxy group, an ethoxy group, and a butoxy group; a dialkylamino group such as a dimethylamino group and a diethylamino group; and a diarylamino group such as a dibenzylamino group and a diphenylamino group.

[0054] Next, a main specific example of a compound represented by the general formula (V) is shown in Table 7 below; however, a compound represented by the general formula (V) is not limited thereto.

[0055]

[Table 7]

* Ortho-position with respect to amino group

[0056]

When the above-mentioned aromatic amine compounds are used to form the interface layer, the aromatic amine compounds may be further mixed with each other. As an organic EL element of the present invention having the interface layer made from the aromatic amine compound, layer structures as shown below can be given:

[0057]

[Table 8]

an anode/ an organic light emitting layer/ an interface layer/ and a cathode;

an anode/ an organic light emitting layer made from a high molecule/ an interface layer/ and a cathode;

an anode/ an organic light emitting layer dispersed in a high molecule/ an interface layer/ and a cathode;

an anode/ a hole transporting layer/ an organic light emitting layer having an electron transporting property/ an interface layer/ and a cathode;

an anode/ an organic light emitting layer having a hole transporting property/ an organic electron transporting layer/ an interface layer/ and a cathode;

an anode/ a hole transporting layer/ an organic light emitting layer having an electron transporting property/ an interface layer/ and a cathode; and

an anode/ a hole transporting layer/ an organic light emitting layer having an electron transporting property/ an electron transporting layer/ an interface layer/ and a cathode.

The thickness of the interface layer 4 is generally set to be 20 to 1,000 Å, and preferably, 50 to 300 Å.

[0058] In the present invention, by using an aromatic diamine compound as a material of an interface layer sandwiched between an organic light emitting layer and a cathode of an organic EL element as shown above, excellent element stability can be achieved. Note that it is possible to employ a reversed structure of FIG 1, and that is, the conductive layer 2b, the interface layer 4, the organic light emitting layer 3, and the conductive layer 2a may be sequentially stacked over the substrate. As described above, the organic EL element of the present invention can be provided between two substrates in which at least one of the two substrates has a high light transmitting property. Similarly, the structures as shown in FIG 2 and FIG 3 can be reversely stacked.

[0059]

[Embodiment] Next, the present invention will be described in more detail in the embodiments; however, the present invention is not limited to descriptions of the embodiments without departing from the scope of the present invention.

Embodiment 1

An organic EL element having the structure shown in FIG 2 was manufactured in the following manner.

[0060] A glass substrate over which an indium tin oxide (ITO) transparent conductive film was deposited with a thickness of 1,200 Å was subjected to ultrasonic cleaning with acetone, and then was washed with pure water. The glass substrate was further subjected to ultrasonic cleaning with isopropyl alcohol, dried with dry nitrogen, and then, subjected to UV/ozone cleaning. Thereafter, the glass substrate was put in a vacuum evaporation apparatus and the vacuum evaporation apparatus was evacuated by using an oil diffusion pump such that the vacuum degree inside the apparatus was set to be 2 x 10^{-6} Torr or less.

[0061] As a material of an organic hole transporting layer, the aromatic amine compound (2) shown in the Table 1

[0062]

[Chemical formula 7] (2)

[0063] was put in a ceramic crucible, and the crucible was heated with a tantalum wire

heater around the crucible to perform evaporation. The temperature of the crucible at this time was controlled in a range of 160 to 170°C. The vacuum degree in evaporation was 2×10^{-6} Torr and an evaporation time was set to be 3 minutes 20 seconds so as to obtain an organic hole transporting layer 3a having a thickness of 600 Å. Subsequently, as a material of an organic electron transporting layer 3b, an 8-hydroxyquinoline complex of aluminum and $Al(C_9H_6NO)_3$ (E1) were evaporated over the organic hole transporting layer 3a in the same manner. At this moment, the temperature of a crucible was controlled in a range of 230 to 270°C. The vacuum degree in evaporation was 2×10^{-6} Torr, the evaporation time was 3 minutes 30 seconds, and a film thickness was 750 Å. This layer served as a light emitting layer.

[0064] Next, as an interface layer 4, the above described aromatic amine compound (2) was evaporated over the organic light emitting layer 3b having an electron transporting property in the same manner as the organic hole transporting layer 3a. The vacuum degree in evaporation was 2×10^{-6} Torr, the evaporation time was 1 minute, and a film thickness was 150 Å. Finally, as a cathode, an alloy electrode including magnesium and silver was formed with a thickness of 1,500 Å by binary co-evaporation. When molybdenum boat was used for evaporation, the vacuum degree was 3×10^{-6} Torr, and the evaporation time was 4 minutes 30 seconds, a glossy film was obtained. An atom ratio between magnesium and silver was set to be 10 : 1.5.

[0065] Results of light emitting characteristics of the organic EL element manufactured in such a manner, which were measured by applying direct voltage so that the ITO electrode (an anode) is positive and the magnesium-silver alloy electrode (a cathode) is negative, were shown in Table 8. This element showed uniform yellow-green light emission, and had a peak wavelength of 560 nm of light emission. Results of light emitting characteristics, which were measured after the element was kept in vacuum, are shown in Table 8. As an area of dark spots, after taking light emission from the element by a CCD camera, the light emission was quantified by image analysis to obtain a value of the area of the dark spots.

[0066] Comparative Example 1

An organic EL element having the structure as shown in FIG. 2 was manufactured

in the same manner as Embodiment 1 with the exception that the interface layer 4 was not provided. Measurement results of light emitting characteristics of the element after manufacturing the element and after keeping it in vacuum were shown in Table 8. It was obvious that dark spots were generated.

[0067]

[Table 9]: Table 8

Element	Measurement condition	Light emitting efficiency [lm/W]	V100 [V]	Dark spot [%]
Embodiment 1	Immediately after manufacturing	0.98	12	0.0
	62 days after	0.78	13	1.2
Comparative Example 1	Immediately after manufacturing	1.14	11	0.0
	62 days after	0.90	14	40.0

• light emitting efficiency is a value by practical luminance (100 cd/m^2).

• V_{100} = voltage by which luminance becomes 100 [cd/m^2].

[0068] Embodiment 2

An element was manufactured in the same manner as Embodiment 1 with the exception that an interface layer 4 was formed using the following aromatic amine compound (24) shown in the Table 4 as substitute for the aromatic amine compound (2) and a thickness of the interface layer was set to be 100 Å. Lihgt emitting characteristics of this element were shown in Table 9.

[0069]

[Chemical Formula 8] (24)

[0070]

[Table 10]: Table 9

Element	Measurement condition	Light emitting efficiency [lm/W]	V100 [V]	Dark spot [%]
Embodiment 2	Immediately after manufacturing	1.11	9	0.0
	62 days after	1.17	9	5.0

[0071]

[Effect of the Invention] According to an organic EL element of the present invention, since an anode, an organic light emitting layer, an interface layer, and a cathode are sequentially stacked over a substrate and a specific compound is used for the interface layer, when voltage is applied to both conductive layers as the electrodes, stable light emitting characteristics can be obtained for a long period.

[0072] Therefore, it is thought that an EL element of the present invention is applicable to a field of a flat panel display (for example, a display for an OA computer or a wall-hanging television), a light source, which utilizes a feature as a plane light emitting body (for example, a light source of a copying machine and a backlight source for a liquid crystal display or instruments), a sign board, and a marker light, and a technical value of the EL element is high.

[Brief Description of the Drawings]

[FIG. 1]: FIG. 1 is a schematic cross sectional view showing one example of an organic EL element of the present invention.

[FIG. 2]: FIG. 2 is a schematic cross sectional view showing another example of an organic EL element of the present invention.

[FIG. 3]: FIG. 3 is a schematic cross sectional view showing another example of an organic EL element of the present invention.

[Description of Reference Numerals]

Reference numeral 1 represents substrate, reference numerals 2a and 2b represent conductive layers, reference numeral 3 represents an organic light emitting layer, reference numeral 3a represents a hole transporting layer, reference numeral 3b represents an organic

electron transporting layer, reference numeral 3c represents an organic electron transporting layer made of a compound different from 3b, and reference numeral 4 represents an interface layer.

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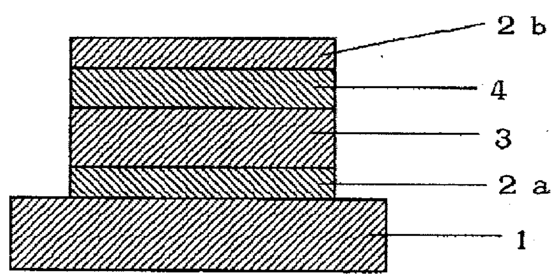
(54)【発明の名称】　有機ＥＬ素子

(57)【要約】
【目的】　長期にわたって安定に駆動させることができる有機ＥＬ素子を提供する。
【構成】　陽極、有機発光層、陰極を順次積層した有機ＥＬ素子であって、前記有機発光層と前記陰極の間に芳香族アミン化合物から成る界面層を設けたことを特徴とする有機ＥＬ素子。
【効果】　低い駆動電圧で実用上十分な輝度の発光を得ることができ、長期間安定した発光性能を示す。



【特許請求の範囲】
【請求項１】　陽極、有機発光層、陰極を順次積層した有機ＥＬ素子であって、前記有機発光層と前記陰極の間に芳香族アミン化合物から成る界面層を設けたことを特徴とする有機ＥＬ素子。
【発明の詳細な説明】
【０００１】
【産業上の利用分野】本発明は有機ＥＬ素子に関するものであり、詳しくは、有機化合物から成る発光層に電界をかけて光を放出する薄膜型デバイスに関するものである。
【０００２】
【従来の技術】従来、薄膜型の電界発光（ＥＬ）素子としては、無機材料のII－VI族化合物半導体であるＺｎＳ、ＣａＳ、ＳｒＳ等に、発光中心であるMnや希土類元素（Ｅｕ、Ｃｅ、Ｔｂ、Sm等）をドープしたものが一般的であるが、上記の無機材料から作製したＥＬ素子は、
１）交流駆動が必要（５０～１０００Ｈｚ）、
２）駆動電圧が高い（～２００Ｖ）、
３）フルカラー化が困難（特に青色が問題）、
４）周辺駆動回路のコストが高い、
という問題点を有している。
【０００３】しかし、近年、上記問題点の改良のため、有機薄膜を用いたＥＬ素子の開発が行われるようになった。特に、発光効率を高めるために電極からのキャリアー注入の効率向上を目的とした電極種類の最適化を行い、芳香族ジアミンから成る有機正孔輸送層と８－ヒドロキシキノリンのアルミニウム錯体から成る発光層を設けた有機ＥＬ素子の開発（Ａｐｐｌ．Ｐｈｙｓ．Ｌｅｔｔ．，５１巻，９１３頁，１９８７年）により、従来のアントラセン等の単結晶を用いたＥＬ素子と比較して発光効率の大幅な改善がなされている。
【０００４】また、有機発光層としては、ポリ（ｐ－フェニレンビニレン）（Ｎａｔｕｒｅ，３４７巻，５３９頁，１９９０年；Ａｐｐｌ．Ｐｈｙｓ．Ｌｅｔｔ．，６１巻，２７９３頁，１９９２年）、ポリ［２－メトキシ，５－（２’－エチルヘキソキシ）－１，４－フェニレンビニレン］（Ａｐｐｌ．Ｐｈｙｓ．Ｌｅｔｔ．，５８巻，１９８２頁，１９９１年；Ｔｈｉｎ　Ｓｏｌｉｄ　Ｆｉｌｍｓ，２１６巻，９６頁，１９９２年；Ｎａｔｕｒｅ，３５７巻，４７７頁，１９９２年）、ポリ（３－アルキルチオフェン）（Ｊｐｎ．Ｊ．Ａｐｐｌ．Ｐｈｙｓ，３０巻，Ｌ１９３８頁，１９９１年；Ｊ．Ａｐｐｌ．Ｐｈｙｓ．，７２巻，５６４頁，１９９２年）等の高分子材料の開発や、ポリビニルカルバゾール等の高分子に発光材料と電子移動材料を混合した素子（応用物理，６１巻，１０４４頁，１９９２年）も開発されている。
【０００５】以上に示した様な有機ＥＬ素子においては、通常、陽極としてはインジウム錫酸化物（ＩＴＯ）のような透明電極が用いられるが、陰極に関しては電子注入を効率よく行うために仕事関数の低い金属電極が用いられ、マグネシウム合金やカルシウム等が使われている。有機ＥＬ素子の最大の問題点は素子の寿命であり、寿命を制限する一つの要因が陰極材料に由来するダークスポット（素子の発光部で発光しない部分をさす）の発生である。このために、長期間保存する場合に、有機ＥＬ素子内のダークスポットの数・大きさが増加して、結果として素子の寿命が短いものとなっている。
【０００６】
【発明が解決しようとする課題】これまでに開示されている有機ＥＬ素子では、ＥＬ発光は陽極から注入された正孔と陰極から注入された電子との再結合によりもたらされる。一般に、キャリアの注入は、電子の場合、陰極と有機発光層との界面における注入障壁を乗り越えて行われなければならない。この電子注入障壁を低くして注入効率を向上させるために、マグネシウム合金やカルシウム等の低仕事関数の金属電極が陰極として使用されている。しかしながら、これらの金属材料は有機発光層との密着性が悪いために有機発光層から剥離したり、陰極を有機発光層上に蒸着する際や蒸着後にも、陰極材料と有機発光層材料との間で反応がおき、長期間の保存中に素子の発光特性の劣化やダークスポットの発生をもたらす。
【０００７】本発明者等は上記実状に鑑み、長期間にワタって安定な発光特性を維持でき、ダークスポットの発生を抑制することができる有機ＥＬ素子を提供することを目的として鋭意検討した結果、有機発光層と陰極の間に芳香族アミン化合物から成る界面層を設けることが好適であることを見い出し、本発明を完成するに至った。
【０００８】
【課題を解決するための手段】すなわち、本発明の要旨は、陽極、有機発光層、陰極を順次積層した有機ＥＬ素子であって、前記有機発光層と前記陰極の間に芳香族アミン化合物から成る界面層を設けたことを特徴とする有機ＥＬ素子に存する。以下、本発明の有機ＥＬ素子について添付図面に従い説明する。
【０００９】図１は本発明の有機ＥＬ素子の構造例を模式的に示す断面図であり、１は基板、２ａ、２ｂは導電層、３は有機発光層、４は界面層を各々表わす。基板１は本発明の有機ＥＬ素子の支持体となるものであり、石英やガラスの板、金属板や金属箔、プラスチックフィルムやシートなどが用いられるが、ガラス板や、ポリエステル、ポリメタアクリレート、ポリカーボネート、ポリサルホンなどの透明な合成樹脂基板が好ましい。
【００１０】基板１上には導電層２ａが設けられるが、この導電層２ａとしては通常、アルミニウム、金、銀、ニッケル、パラジウム、テルル等の金属、インジウム及び／またはスズの酸化物などの金属酸化物やヨウ化銅、

カーボンブラック、あるいは、ポリ（３－メチルチオフェン）等の導電性高分子などにより構成される。導電層の形成は通常、スパッタリング法、真空蒸着法などにより行われることが多いが、銀などの金属微粒子あるいはヨウ化銅、カーボンブラック、導電性の金属酸化物微粒子、導電性高分子微粉末などの場合には、適当なバインダー樹脂溶液に分散し、基板上に塗布することにより形成することもできる。さらに、導電性高分子の場合は電界重合により直接基板上に薄膜を形成したり、基板上に塗布して形成することもできる（Ａｐｐｌ．Ｐｈｙｓ．Ｌｅｔｔ．，６０巻，２７１１頁，１９９２年）。上記の導電層は異なる物質で積層することも可能である。導電層２ａの厚みは、必要とする透明性により異なるが、透明性が必要とされる場合は、可視光の透過率が６０％以上、好ましくは８０％以上透過することが望ましく、この場合、厚みは、通常、５０～１００００Å、好ましくは１００～５０００Å程度である。

【００１１】不透明でよい場合は導電層２ａは基板１と同一でもよい。また、さらには上記の導電層を異なる物質で積層することも可能である。図１の例では、導電層２ａは陽極（アノード）として正孔注入の役割を果たすものである。一方、導電層２ｂは陰極（カソード）として、界面層４を通して有機発光層３に電子を注入する役割を果たす。導電層２ｂとして用いられる材料は、前記導電層２ａ用の材料を用いることが可能であるが、効率よく電子注入を行なうには、仕事関数の低い金属が好ましく、スズ、マグネシウム、インジウム、アルミニウム、銀等の適当な金属またはそれらの合金が用いられる。導電層２ｂの膜厚は通常、導電層２ａと同様である。また、図１には示してはいないが、導電層２ｂの上にさらに基板１と同様の基板を設けることもできる。但し、導電層２ａと２ｂの少なくとも一方は透明性の良いことがＥＬ素子としては必要である。このことから、導電層２ａと２ｂの一方は、１００～５０００Åの膜厚であることが好ましく、透明性の良いことが望まれる。

【００１２】導電層２ａの上には有機発光層３が設けられるが、有機発光層３は、電界を与えられた電極間において、陽極から注入された正孔と陰極から注入された電子がを効率よく輸送して再結合させ、かつ、再結合により効率よく発光する材料から形成される。通常、この有機発光層３は発光効率の向上のために、図２に示すように、正孔輸送層３ａと電子輸送層３ｂに分割して機能分離型にすることが行われる（Ａｐｐｌ．Ｐｈｙｓ．Ｌｅｔｔ．，５１巻，９１３頁，１９８７年）。

【００１３】上記の機能分離型素子において、正孔輸送材料としては、導電層２ａからの正孔注入効率が高く、かつ、注入された正孔を効率よく輸送することができる材料であることが必要である。そのためには、イオン化ポテンシャルが小さく、しかも正孔移動度が大きく、さらに安定性にすぐれ、トラップとなる不純物が製造時や使用時に発生しにくいことが要求される。

【００１４】このような正孔輸送化合物としては、例えば、特開昭５９－１９４３９３号公報及び米国特許第４，１７５，９６０号の第１３～１４欄に解説される、Ｎ，Ｎ’－ジフェニル－Ｎ，Ｎ’－（３－メチルフェニル）－１，１’－ビフェニル－４，４’－ジアミン：１，１’－ビス（４－ジ－ｐ－トリルアミノフェニル）シクロヘキサン：４，４’－ビス（ジフェニルアミノ）クワドロフェニルなどの芳香族アミン系化合物、特開平２－３１１５９１号公報に示されるヒドラゾン化合物、米国特許第４，９５０，９５０号公報に示されるシラザン化合物等が挙げられる。これらの化合物は、単独で用いるか、必要に応じて、各々、混合して用いてもよい。上記の化合物以外に、ポリビニルカルバゾールやポリシラン（Ａｐｐｌ．Ｐｈｙｓ．Ｌｅｔｔ．，５９巻，２７６０頁，１９９１年）等の高分子材料が挙げられる。

【００１５】上記の有機正孔輸送材料は塗布法あるいは真空蒸着法により前記導電層２ａ上に積層することにより正孔輸送層３ａを形成する。塗布の場合は、有機正孔輸送化合物を１種または２種以上と必要により正孔のトラップにならないバインダー樹脂や、レベリング剤等の塗布性改良剤などの添加剤を添加し溶解した塗布溶液を調整し、スピンコート法などの方法により導電層２ａ上に塗布し、乾燥して有機正孔輸送層３を形成する。バインダー樹脂としては、ポリカーボネート、ポリアリレート、ポリエステル等が挙げられる。バインダー樹脂は添加量が多いと正孔移動度を低下させるので、少ない方が望ましく、５０重量％以下が好ましい。

【００１６】真空蒸着法の場合には、有機正孔輸送材料を真空容器内に設置されたルツボに入れ、真空容器内を適当な真空ポンプで１０^{-6}Ｔｏｒｒにまで排気した後、ルツボを加熱して、正孔輸送材料を蒸発させ、ルツボと向き合って置かれた基板上に層を形成する。正孔輸送層３ａの膜厚は、通常、１００～３０００Å、好ましくは３００～１０００Åである。この様に薄い膜を一様に形成するためには、真空蒸着法がよく用いられる。

【００１７】正孔輸送層３ａの材料としては有機化合物の代わりに無機材料を使用することも可能である。無機材料に要求される条件は、有機正孔輸送化合物と同じである。正孔輸送層３に用いられる無機材料としては、ｐ型水素化非晶質シリコン、ｐ型水素化非晶質炭化シリコン、ｐ型水素化微結晶性炭化シリコン、あるいは、ｐ型硫化亜鉛、ｐ型セレン化亜鉛等が挙げられる。これらの無機正孔輸送層はＣＶＤ法、プラズマＣＶＤ法、真空蒸着法、スパッタ法等により形成される。

【００１８】無機正孔輸送層の膜厚も有機正孔輸送層と同様に、通常、１００～３０００Å、好ましくは３００～１０００Åである。正孔輸送層３ａの上には電子輸送層３ｂが設けられるが、電子輸送層３ｂは、電界を与えられた電極間において陰極からの電子を効率よく正孔輸

送層の方向に輸送することができる化合物より形成される。

【0019】有機電子輸送化合物としては、導電層２ｂからの電子注入効率が高く、かつ、注入された電子を効率よく輸送することができる化合物であることが必要である。そのためには、電子親和力が大きく、しかも電子移動度が大きく、さらに安定性にすぐれトラップとなる不純物が製造時や使用時に発生しにくい化合物であることが要求される。

【0020】このような条件を満たす材料としては、テトラフェニルブタジエンなどの芳香族化合物（特開昭５７－５１７８１号公報）、８－ヒドロキシキノリンのアルミニウム錯体などの金属錯体（特開昭５９－１９４３９３号公報）、シクロペンタジエン誘導体（特開平２－２８９６７５号公報）、ペリノン誘導体（特開平２－２８９６７６号公報）、オキサジアゾール誘導体（特開平２－２１６７９１号公報）、ビススチリルベンゼン誘導体（特開平１－２４５０８７号公報、同２－２２２４８４号公報）、ペリレン誘導体（特開平２－１８９８９０号公報、同３－７９１号公報）、クマリン化合物（特開平２－１９１６９４号公報、同３－７９２号公報）、希土類錯体（特開平１－２５６５８４号公報）、ジスチリルピラジン誘導体（特開平２－２５２７９３号公報）、ｐ－フェニレン化合物（特開平３－３３１８３号公報）、チアジアゾロピリジン誘導体（特開平３－３７２９２号公報）、ピロロピリジン誘導体（特開平３－３７２９３号公報）、ナフチリジン誘導体（特開平３－２０３９８２号公報）などが挙げられる。

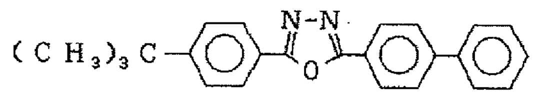
【0021】これらの化合物を用いた有機電子輸送層は、電子を輸送する役割と、正孔と電子の再結合の際に発光をもたらす役割を同時に果しており、発光層を兼ねている。有機正孔輸送化合物が発光機能を有する場合は、有機電子輸送層は電子を輸送する役割だけを果たす。

【0022】素子の発光効率を向上させるとともに発光色を変える目的で、例えば、８－ヒドロキシキノリンのアルミニウム錯体をホスト材料として、クマリン等のレーザ用蛍光色素をドープすること（Ｊ．Ａｐｐｌ．Ｐｈｙｓ．，６５巻，３６１０頁，１９８９年）も行われている。本発明においても上記の有機電子輸送材料をホスト材料として各種の蛍光色素を10^{-3}～１０モル％ドープすることにより、素子の発光特性をさらに向上させることができる。電子輸送層３ｂの膜厚は、通常、１００～２０００Å、好ましくは３００～１０００Åである。

【0023】有機電子輸送層も有機正孔輸送層と同様の方法で形成することができるが、通常は真空蒸着法が用いられる。有機ＥＬ素子の発光効率をさらに向上させる方法として、電子輸送層３ｂの上にさらに他の電子輸送層３ｃを積層することが考えられる（図３参照）。この電子輸送層３ｃに用いられる化合物には、陰極からの電子注入が容易で、電子の輸送能力がさらに大きいことが要求される。この様な電子輸送材料としては、

【0024】

【化１】



【0025】などのオキサジアゾール誘導体（Ａｐｐｌ．Ｐｈｙｓ．Ｌｅｔｔ．，５５巻，１４８９頁，１９８９年）やそれらをＰＭＭＡ等の樹脂に分散した系（Ａｐｐｌ．Ｐｈｙｓ．Ｌｅｔｔ．，６１巻，２７９３頁，１９９２年）、または、ｎ型水素化非晶質炭化シリコン、ｎ型硫化亜鉛、ｎ型セレン化亜鉛等が挙げられる。電子輸送層３ｃの膜厚は、通常、５０～２０００Å、好ましくは１００～１０００Åである。

【0026】また、機能分離を行わない単層型の有機発光層３としては、先に挙げたポリ（ｐ－フェニレンビニレン）（Ｎａｔｕｒｅ，３４７巻，５３９頁，１９９０年；Ａｐｐｌ．Ｐｈｙｓ．Ｌｅｔｔ．，６１巻，２７９３頁，１９９２年）、ポリ［２－メトキシ，５－（２’－エチルヘキソキシ）－１，４－フェニレンビニレン］（Ａｐｐｌ．Ｐｈｙｓ．Ｌｅｔｔ．，５８巻，１９８２頁，１９９１年；Ｔｈｉｎ Ｓｏｌｉｄ Ｆｉｌｍｓ，２１６巻，９６頁，１９９２年；Ｎａｔｕｒｅ，３５７巻，４７７頁，１９９２年）、ポリ（３－アルキルチオフェン）（Ｊｐｎ．Ｊ．Ａｐｐｌ．Ｐｈｙｓ，３０巻，Ｌ１９３８頁，１９９１年；Ｊ．Ａｐｐｌ．Ｐｈｙｓ．，７２巻，５６４頁，１９９２年）等の高分子材料や、ポリビニルカルバゾール等の高分子に発光材料と電子移動材料を混合した系（応用物理，６１巻，１０４４頁，１９９２年）がある。

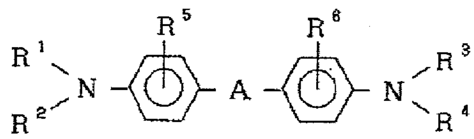
【0027】有機発光層の上に界面層４が設けられる。界面層の役割としては、有機発光層との親和性があると同時に陰極との密着性がよく、かつ、化学的に安定で陰極形成時及び／または形成後の有機発光層と陰極の反応を抑制する効果を有することが挙げられる。また、均一な薄膜形状を与えることも陰極との密着性の点で重要である。このような役割を果たす材料として、芳香族アミン化合物が好適であることを本発明者は見い出した。

【0028】芳香族アミン化合物から成る界面層は、前述の有機正孔輸送材料と同様に、塗布法あるいは真空蒸着法により形成される。本発明においては、以上の様に、有機ＥＬ素子における有機発光層／陰極間の界面層材料として、芳香族アミン化合物を用いることにより、優れた素子の安定性が達成される。

【0029】芳香族アミン化合物として、好適なものとしては下記一般式（Ｉ）～（Ｖ）のものが挙げられる。

【0030】

【化２】

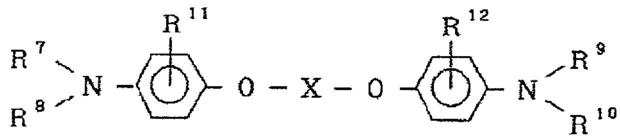


（Ｉ）

【００３１】（式中、Ａは置換基を有していてもよい２価の炭化水素残基または直接結合を示し、R^1、R^2、R^3およびR^4はそれぞれ置換もしくは未置換の、アルキル基、シクロアルキル基、アルケニル基、置換もしくは未置換のアリール基、またはアラルキル基を表し、これらは同一でも異なっていてもよく、R^5およびR^6はそれぞれ水素原子、ハロゲン原子、水酸基、置換もしくは未置換の、アルキル基、シクロアルキル基、アルケニル基、アリール基、アラルキル基、アルコキシ基、ジアルキルアミノ基、またはジアリールアミノ基を表す。）

【００３２】

【化３】

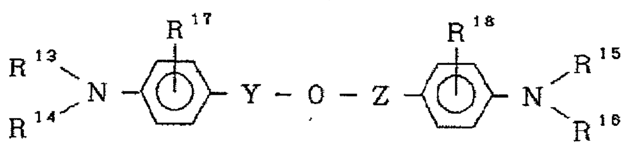


（II）

【００３３】（式中、R^7、R^8、R^9およびR^{10}はそれぞれ置換もしくは未置換の、アルキル基、シクロアルキル基、アルケニル基、置換もしくは未置換のアリール基、またはアラルキル基を表し、これらは同一でも異なっていてもよく、R^{11}およびR^{12}は、各々、水素原子、ハロゲン原子、もしくは、水酸基、または、各々置換基を有していてもよい、飽和あるいは不飽和の脂肪族炭化水素基、芳香族炭化水素基、アルコキシ基、アリールオキシ基、ジアルキルアミノ基、もしくは、ジアリールアミノ基を表し、Ｘは置換基を有していてもよい２価の炭化水素残基を表す。）

【００３４】

【化４】

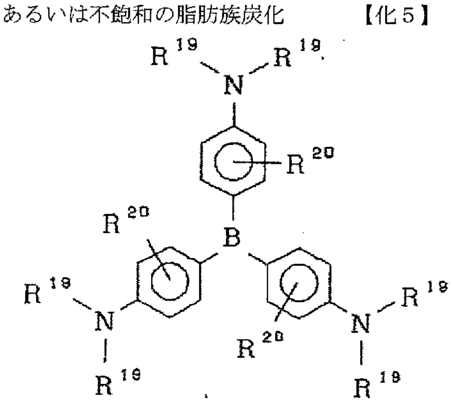


（III）

【００３５】（式中、R^{13}、R^{14}、R^{15}およびR^{16}はそれぞれ置換もしくは未置換の、アルキル基、シクロアルキル基、アルケニル基、置換もしくは未置換のアリール基、またはアラルキル基を表し、これらは同一でも異なっていてもよく、R^{17}およびR^{18}は、各々、水素原子、ハロゲン原子、もしくは、水酸基、または、各々置換基を有していてもよい、飽和あるいは不飽和の脂肪族炭化水素基、芳香族炭化水素基、アルコキシ基、アリールオキシ基、ジアルキルアミノ基、もしくは、ジアリールアミノ基を表し、ＹおよびＺは置換基を有していてもよい２価の炭化水素残基または直接結合を表し、これらは同一でも異なっていてもよい。）

【００３６】

【化５】



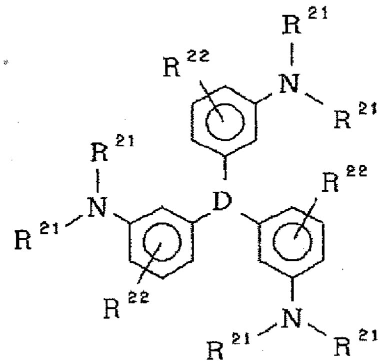
（IV）

【００３７】（式中、Ｂは置換基を有していてもよい３価の炭化水素残基または窒素原子を示し、R^{19}は置換もしくは未置換の、アルキル基、シクロアルキル基、アルケニル基、置換もしくは未置換のアリール基、またはアラルキル基を表し、これらは同一でも異なっていてもよく、R^{20}は水素原子、ハロゲン原子、水酸基、置換もしくは未置換の、アルキル基、シクロアルキル基、アルケニル基、アリール基、アラルキル基、アルコキシ基、ジ

アルキルアミノ基、またはジアリールアミノ基を表す。）

【0038】

【化6】



（V）

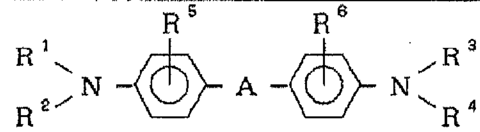
【0039】（式中、Dは置換基を有していてもよい3価の炭化水素残基または窒素原子を示し、R^{21}は置換もしくは未置換の、アルキル基、シクロアルキル基、アルケニル基、置換もしくは未置換のアリール基、またはアラルキル基を表し、これらは同一でも異なっていてもよく、R^{22}は水素原子、ハロゲン原子、水酸基、置換もしくは未置換の、アルキル基、シクロアルキル基、アルケニル基、アリール基、アラルキル基、アルコキシ基、ジアルキルアミノ基、またはジアリールアミノ基を表す。）前記一般式（I）において、好ましくは、Aは$-CH_2-$、$-CH_2-CH_2-$、$-CH_2CH_2CH_2-$ 等の置換基を有していてもよいアルキレン基；シクロヘキシレン基等のシクロアルキレン基；$-CH=CH-$、$-C(CH3)=CH-$、$-CH_2-CH=CH-CH_2-$、$-CH=CH-CH=CH-$等の置換基を有していてもよいアルケニレン基；置換基を有していてもよいフェニレン基、ナフチレン基、フェナンスレン基等のアリーレン基または直接結合を示し、R^1、R^2、R^3およびR^{4}はそれぞれ置換基を有していてもよいメチル基、エチル基、プロピル基、ブチル基、ヘキシル基等のアルキル基；シクロヘキシル基等のシクロアルキル基；ビニル基等のアルケニル基；アルキル基、アルコキシ基、ハロゲン原子、アミノ基、アリール基等の置換基を有していてもよいフェニル基、ナフチル基等のアリール基；ベンジル基、ナフチルメチル基、フェネチル基等のアラルキル基を表し、これらは同一でも異なっていてもよく、R^5およびR^6はそれぞれ水素原子、ハロゲン原子、水酸基、置換基を有していてもよいメチル基、エチル基、プロピル基、ブチル基、ヘキシル基等のアルキル基；シクロヘキシル基等のシクロアルキル基；ビニル基等のアルケニル基；アルキル基、アルコキシ基、ハロゲン原子、アミノ基、アリール基等の置換基を有していてもよいフェニル基、ナフチル基等のアリール基；ベンジル基、ナフチルメチル基、フェネチル基等のアラルキル基；メトキシ基、エトキシ基、ブトキシ基等のアルコキシ基；ジメチルアミノ基、ジエチルアミノ基等のジアルキルアミノ基；ジベンジルアミノ基、ジフェニルアミノ基等のジアリールアミノ基を表し、これらは同一でも異なっていてもよい。

【0040】次に前記一般式（I）で示される化合物の主な具体例を以下の表－1および表－2に示すが、これらに限定するものではない。

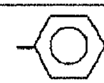
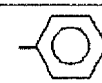
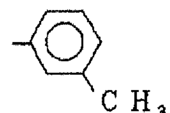
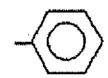
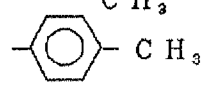
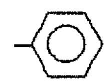
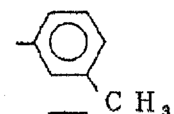
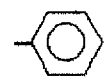
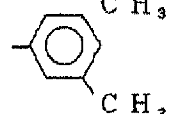
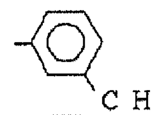
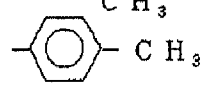
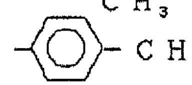
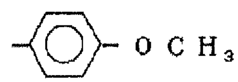
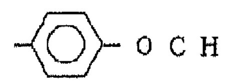
【0041】

【表1】

表－1



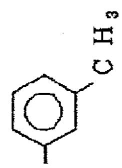
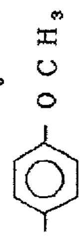
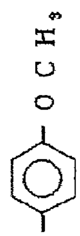
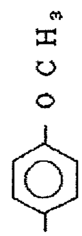
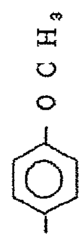
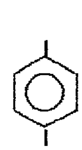
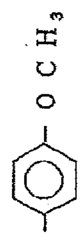
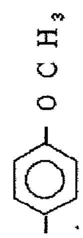
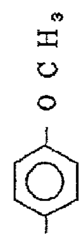
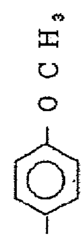
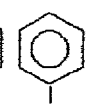
＊アミノ基に対して
オルト位

番号	A	R^1, R^3	R^2, R^4	R^5, R^6
（１）	直接結合			－H
（２）	直接結合			－H
（３）	直接結合			－H
（４）	直接結合			$-CH_3$＊
（５）	直接結合			－H
（６）	直接結合			$-CH_3$＊
（７）	直接結合			$-CH_3$＊

【００４２】

【表２】

表－2

番号	A	R^1, R^3	R^2, R^4	R^5, R^6
(8)	直接結合	$-CH_3$	$-CH_3$	$-H$
(9)	$-CH_2-$			$-H$
(10)	$-CH_2-$			$-H$
(11)				$-H$
(12)				$-H$
(13)	$-CH=CH-$			$-H$
(14)	$-CH=CH-$			$-H$
(15)	$-(CH=CH-)_3-$			$-H$

【００４３】前記一般式（II）において、好ましくは、R^7、R^8、R^9 およびR^{10}はそれぞれ置換基を有していてもよいメチル基、エチル基、プロピル基、ブチル基、ヘキシル基等のアルキル基；シクロヘキシル基等のシクロアルキル基；アリル基等のアルケニル基；アルキル基、アルコキシ基、ハロゲン原子、アミノ基、アリール基等の置換基を有していてもよいフェニル基、ナフチル基等のアリール基；ベンジル基、ナフチルメチル基、フェネチル基等のアラルキル基を表し、これらは同一でも異なっていてもよく、R^{11}およびR^{12}はそれぞれ水素原子、ハロゲン原子、水酸基、置換基を有していてもよいメチル基、エチル基、プロピル基、ブチル基、ヘキシル基等のアルキル基；シクロヘキシル基等のシクロアルキル基；ビニル基等のアルケニル基；アルキル基、アルコキシ基、ハロゲン原子、アミノ基、アリール基等の置換基を有していてもよいフェニル基、ナフチル基等のアリール基；ベンジル基、ナフチルメチル基、フェネチル基等のアラルキル基；メトキシ基、エトキシ基、ブトキシ基等のアルコキシ基；ジメチルアミノ基、ジエチルアミノ基等のジアルキルアミノ基；ジベンジルアミノ基、ジフェニルアミノ基等のジアリールアミノ基を表し、これらは同一でも異なっていてもよい。Xはメチレン基、プロピレン基、キシリレン基、シクロヘキシレン基、ビニレン基、フェニレン基、－CO－などの２価の炭化水素残基を表し、これらは、ハロゲン原子、水酸基、飽和あるいは不飽和の炭化水素基、アルコキシ基、アリールオキシ基、ジアルキルアミノ基、ジアリールアミノ基などの置換基を有していてもよい。

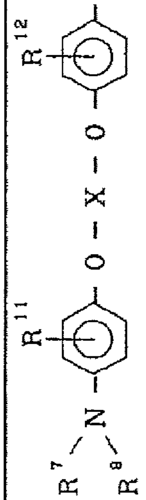
【００４４】次に前記一般式（II）で示される化合物の主な具体例を以下の表－３および表－４に示すが、これ

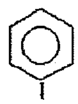
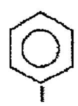
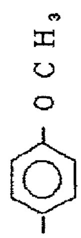
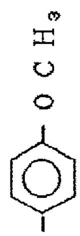
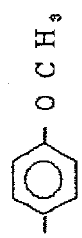
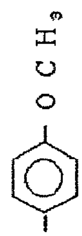
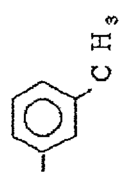
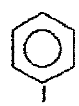
らに限定するものではない。

【００４５】

【表３】

表－３



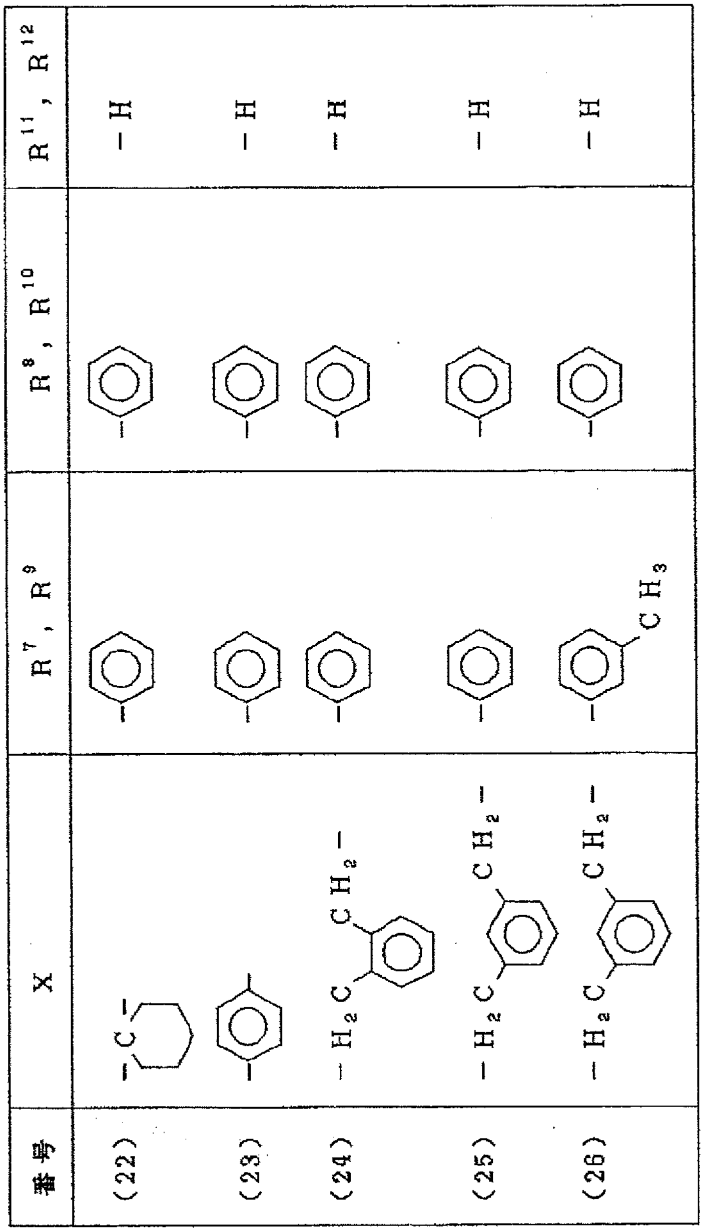
番号	X	R^7, R^9	R^8, R^{10}	R^{11}, R^{12}
(16)	$-C(=O)-$			－H
(17)	$-C(=O)-$			－H
(18)	$-CH_2-$			－H
(19)	$-CH_2-$			－H
(20)	$-CH_2-$			$-CH_3$＊
(21)	$-CH=CH-$			－H

＊アミノ基に対してオルト位

【０　０　４　６】

【表４】

表－4



【0047】前記一般式（III）において、好ましくは、R^{13}、R^{14}、R^{15}およびR^{16}はそれぞれ置換基を有していてもよいメチル基、エチル基、プロピル基、ブチル基、ヘキシル基等のアルキル基；シクロヘキシル基等のシクロアルキル基；アリル基等のアルケニル基；アルキル基、アルコキシ基、ハロゲン原子、アミノ基、アリール基等の置換基を有していてもよいフェニル基、ナフチル基等のアリール基；ベンジル基、ナフチルメチル基、フェネチル基等のアラルキル基を表し、これらは同一でも異なっていてもよく、R^{17}およびR^{18}はそれぞれ水素原子、ハロゲン原子、水酸基、置換基を有していてもよいメチル基、エチル基、プロピル基、ブチル基、ヘキシル基等のアルキル基；シクロヘキシル基等のシクロアルキル基；ビニル基等のアルケニル基；アルキル基、アルコキシ基、ハロゲン原子、アミノ基、アリール基等の置換基を有していてもよいフェニル基、ナフチル基等のアリール基；ベンジル基、ナフチルメチル基、フェネチル基等のアラルキル基；メトキシ基、エトキシ基、ブトキシ基等のアルコキシ基；ジメチルアミノ基、ジエチルアミノ基等のジアルキルアミノ基；ジベンジルアミノ基、ジフェニルアミノ基等のジアリールアミノ基を表し、これらは同一でも異なっていてもよい。YおよびZはメチレン基、プロピレン基、キシリレン基、シクロヘキシレン基、ビニレン基、フェニレン基などの２価の炭化水素残基または直接結合を表し、これらはハロゲン原子、水酸基、飽和あるいは不飽和の炭化水素基、アルコ

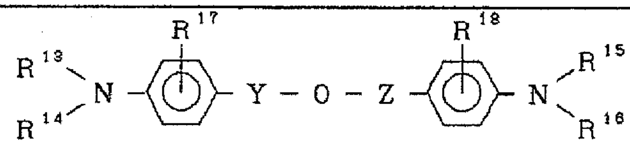
キシ基、アリールオキシ基、ジアルキルアミノ基、ジアリールアミノ基などの置換基を有していてもよい。

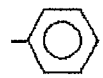
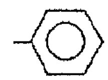
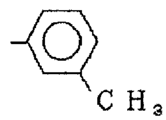
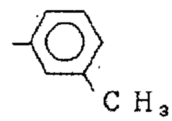
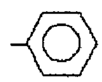
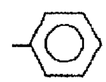
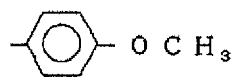
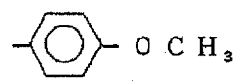
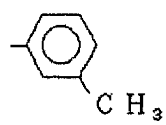
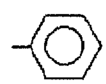
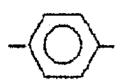
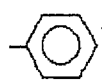
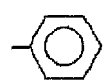
【0048】次に前記一般式（III）で示される化合物の主な具体例を以下の表－５に示すが、これらに限定するものではない。

【0049】

【表5】

表－５



番号	Y，Z	R^{13}，R^{15}	R^{14}，R^{16}	R^{17}，R^{18}
(27)	直接結合			－H
(28)	直接結合			－H
(29)	$-CH_2-$			－H
(30)	$-CH_2-$			－H
(31)	$-CH_2-$			$-CH_3$＊
(32)				－H

＊アミノ基に対してオルト位

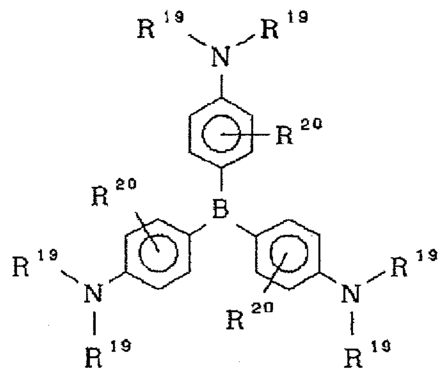
【0050】前記一般式（IV）において、好ましくは、Bは－CH＝；１，３，５の位置の水素原子を除いたベンゼン残基；１，３，５の位置の水素原子を除いたシクロヘキサン残基；窒素原子を示し、R^{19}は、置換基を有していてもよいメチル基、エチル基、プロピル基、ブチル基、ヘキシル基等のアルキル基；シクロヘキシル基等のシクロアルキル基；ビニル基等のアルケニル基；アルキル基、アルコキシ基、ハロゲン原子、アミノ基、アリール基等の置換基を有していてもよいフェニル基、ナフチル基等のアリール基；ベンジル基、ナフチルメチル基、フェネチル基等のアラルキル基を表し、R^{20}は、水素原子、ハロゲン原子、水酸基、置換基を有していてもよいメチル基、エチル基、プロピル基、ブチル基、ヘキシル基等のアルキル基；シクロヘキシル基等のシクロアルキル基；ビニル基等のアルケニル基；アルキル基、アルコキシ基、ハロゲン原子、アミノ基、アリール基等の置換基を有していてもよいフェニル基、ナフチル基等のアリール基；ベンジル基、ナフチルメチル基、フェネチル基等のアラルキル基；メトキシ基、エトキシ基、ブトキシ基等のアルコキシ基；ジメチルアミノ基、ジエチルアミノ基等のジアルキルアミノ基；ジベンジルアミノ基、ジフェニルアミノ基等のジアリールアミノ基を表す。

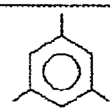
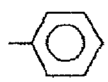
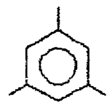
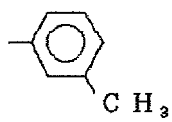
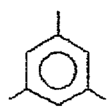
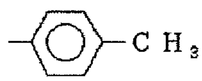
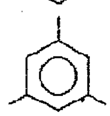
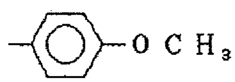
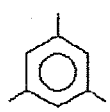
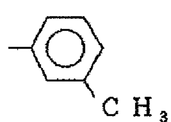
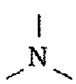
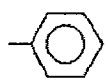
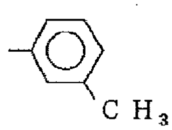
【0051】次に前記一般式（IV）で示される化合物の主な具体例を以下の表－６に示すが、これらに限定するものではない。

【0052】

【表6】

表－6



番号	B	R^{19}	R^{20}
(33)			－H
(34)			－H
(35)			－H
(36)			－H
(37)			$-CH_3$＊
(38)			－H
(39)			－H

＊アミノ基に対してオルト位

【００５３】前記一般式（Ｖ）において、好ましくは、Ｄは－ＣＨ＝；１，３，５の位置の水素原子を除いたベンゼン残基；１，３，５の位置の水素原子を除いたシクロヘキサン残基；窒素原子を示し、R^{21}は、置換基を有していてもよいメチル基、エチル基、プロピル基、ブチル基、ヘキシル基等のアルキル基；シクロヘキシル基等のシクロアルキル基；ビニル基等のアルケニル基；アルキル基、アルコキシ基、ハロゲン原子、アミノ基、アリール基等の置換基を有していてもよいフェニル基、ナフチル基等のアリール基；ベンジル基、ナフチルメチル基、フェネチル基等のアラルキル基を表し、R^{22}は、水素原子、ハロゲン原子、水酸基、置換基を有していてもよいメチル基、エチル基、プロピル基、ブチル基、ヘキシル基等のアルキル基；シクロヘキシル基等のシクロア

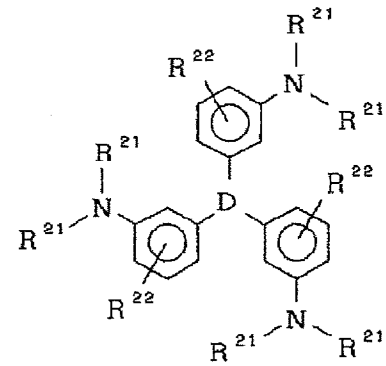
ルキル基；ビニル基等のアルケニル基；アルキル基、アルコキシ基、ハロゲン原子、アミノ基、アリール基等の置換基を有していてもよいフェニル基、ナフチル基等のアリール基；ベンジル基、ナフチルメチル基、フェネチル基等のアラルキル基；メトキシ基、エトキシ基、ブトキシ基等のアルコキシ基；ジメチルアミノ基、ジエチルアミノ基等のジアルキルアミノ基；ジベンジルアミノ基、ジフェニルアミノ基等のジアリールアミノ基を表す。

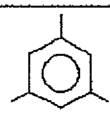
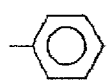
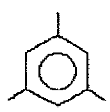
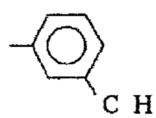
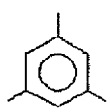
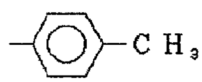
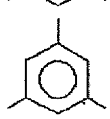
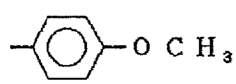
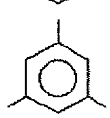
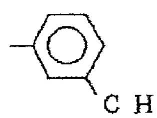
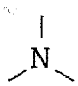
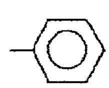
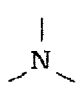
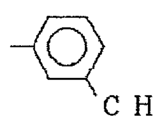
【００５４】次に前記一般式（V）で示される化合物の主な具体例を以下の表－７に示すが、これらに限定するものではない。

【００５５】

【表７】

表－７



番号	D	R^{21}	R^{22}
(40)			－H
(41)			－H
(42)			－H
(43)			－H
(44)			$-CH_3$＊
(45)			－H
(46)			－H

＊アミノ基に対してオルト位

【００５６】上記に示した芳香族アミン化合物を界面層として形成する場合、これらの化合物をさらに混合して用いてもよい。芳香族アミン化合物から成る界面層を有する本発明における有機ＥＬ素子は、以下に示すような層構成のものが挙げられる；

【００５７】

【表８】

陽極／有機発光層／界面層／陰極

陽極／高分子から成る有機発光層／界面層／陰極

陽極／高分子に分散させた有機発光層／界面層／陰極

陽極／正孔輸送層／有機電子輸送性発光層／界面層／陰極

陽極／有機正孔輸送性発光層／有機電子輸送層／界面層／陰極

陽極／正孔輸送層／有機電子輸送性発光層／界面層／陰極

陽極／正孔輸送層／有機電子輸送性発光層／電子輸送層／界面層／陰極

界面層４の膜厚は、通常、２０～１０００Å、好ましくは５０～３００Åである。

【００５８】本発明においては、以上のように、有機ＥＬ素子における有機発光層／陰極間の界面層材料として、芳香族ジアミン化合物を用いることにより、優れた素子の安定性が達成される。尚、図１とは逆の構造、すなわち、基板上に導電層２ｂ、界面層４、有機発光層３、導電層２ａの順に積層することも可能であり、既述した様に少なくとも一方が透明性の高い２枚の基板の間に本発明の有機ＥＬ素子を設けることも可能である。同様に、図２及び図３とは逆の構造に積層することも可能である。

【００５９】

【実施例】次に、本発明を実施例によって更に具体的に説明するが、本発明はその要旨を越えない限り、以下の実施例の記載に限定されるものではない。

実施例１

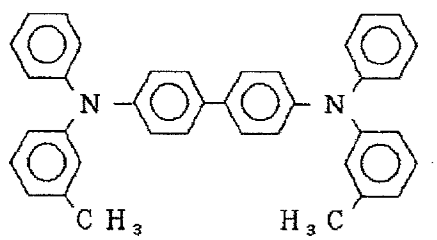
図２に示す構造を有する有機ＥＬ素子を以下の方法で作製した。

【００６０】ガラス基板上にインジウム・スズ酸化物（ＩＴＯ）透明導電膜を１２００Å堆積したものをアセトンで超音波洗浄、純水で水洗、イソプロピルアルコールで超音波洗浄、乾燥窒素で乾燥、ＵＶ／オゾン洗浄を行った後、真空蒸着装置内に設置して、装置内の真空度が2×10^{-6}Torr以下になるまで油拡散ポンプを用いて排気した。

【００６１】有機正孔輸送層材料として、前記表－１に示した芳香族アミン化合物（２）

【００６２】

【化７】



（２）

【００６３】をセラミックるつぼに入れ、るつぼの周囲のタンタル線ヒーターで加熱して蒸着を行った。この時のるつぼの温度は、１６０～１７０℃の範囲で制御した。蒸着時の真空度は2×10^{-6}Ｔｏｒｒで、蒸着時間３分２０秒で膜厚６００Åの有機正孔輸送層３ａを得た。次に、有機電子輸送層３ｂの材料として、アルミニウムの８－ヒドロキシキノリン錯体、$Al(C_9H_6NO)_3$（Ｅ１）を上記有機正孔輸送層３ａの上に同様にして蒸着を行なった。この時のるつぼの温度は２３０～２７０℃の範囲で制御した。蒸着時の真空度は2×10^{-6}Ｔｏｒｒ、蒸着時間は３分３０秒、膜厚は７５０Åであった。この層は発光層としての役割を果たす。

【００６４】次に、界面層４として、上記芳香族アミン化合物（２）を有機電子輸送性発光層３ｂの上に、有機正孔輸送層３ａと同様にして蒸着を行った。蒸着時の真空度は2×10^{-6}Ｔｏｒｒ、蒸着時間は１分、膜厚は１５０Åであった。最後に陰極として、マグネシウムと銀の合金電極を２元同時蒸着法によって膜厚１５００Åで蒸着した。蒸着はモリブデンボートを用いて、真空度は3×10^{-6}Ｔｏｒｒ、蒸着時間は４分３０秒で光沢のある膜が得られた。マグネシウムと銀の原子比は１０：１．５であった。

【００６５】この様にして作製した有機ＥＬ素子のＩＴＯ電極（陽極）にプラス、マグネシウム・銀合金電極（陰極）にマイナスの直流電圧を印加して測定した発光特性の結果を表－８に示す。この素子は一様な黄緑色の発光を示し、発光のピーク波長は５６０ｎｍであった。この素子を真空中に保存後、発光特性を測定した結果を表－８に示す。ダークスポットの面積は、素子からの発光をＣＣＤカメラを用いて撮影した後、画像解析により定量化した値を示す。

【００６６】比較例１

界面層４を設けない他は実施例１と同様にして、図２に示す構造の有機ＥＬ素子を作製した。この素子の作製後と真空中保存後の発光特性の測定結果を表－８に示す。ダークスポットの発生は明かであった。

【００６７】

【表９】

表－８

素　子	測定条件	発光効率 [lm／W]	V_{100} [Ｖ]	ダークスポット [％]
実施例１	作製直後	０．９８	１２	０．０
	６２日後	０．７８	１３	１．２
比較例１	作製直後	１．１４	１１	０．０
	６２日後	０．９０	１４	４０．０

・発光効率は実用輝度（100 cd／m²）での値

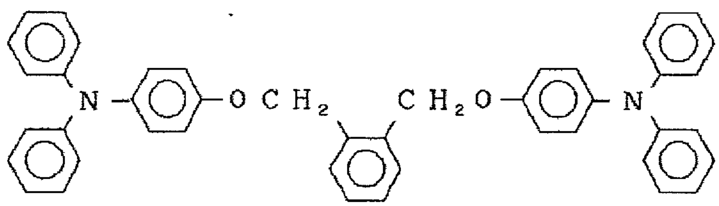
・V_{100}＝輝度が100[cd／m²]となる電圧

【００６８】実施例２

界面層４として、前記表－４に示した下記芳香族アミン化合物（２４）を芳香族アミン化合物（２）の代わりに用いてその膜厚を１００Åとした他は、実施例１と同様にして素子を作製した。この素子の発光特性を表－９に示す。

【００６９】

【化８】



（２４）

【００７０】

【表１０】

表－９

素　子	測定条件	発光効率 [lm／W]	V_{100} [Ｖ]	ダークスポット [％]
実施例２	作製直後	１．１１	９	０．０
	６２日後	１．１７	９	５．０

【００７１】

【発明の効果】本発明の有機ＥＬ素子によれば、陽極（アノード）、有機発光層、界面層、陰極（カソード）、が基板上に順次設けられ、しかも、界面層に特定の化合物を使用しているため、両導電層を電極として電圧を印加した場合、長期に渡り、安定した発光特性を得ることができる。

【００７２】従って、本発明のＥＬ素子はフラットパネル・ディスプレイ（例えばＯＡコンピュータ用や壁掛けテレビ）の分野や面発光体としての特徴を生かした光源（例えば、複写機の光源、液晶ディスプレイや計器類のバックライト光源）、表示板、標識灯への応用が考えられ、その技術的価値は大きいものである。

【図面の簡単な説明】

【図１】本発明の有機ＥＬ素子の一例を示した模式断面図。

【図２】本発明の有機ＥＬ素子の別の例を示した模式断面図。

【図３】本発明の有機ＥＬ素子の別の例を示した模式断面図。

【符号の説明】

１　基板

２ａ、２ｂ　導電層

３　有機発光層

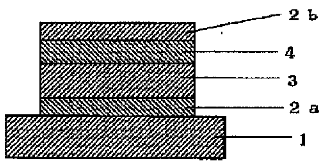
３ａ　正孔輸送層

３ｂ　有機電子輸送層

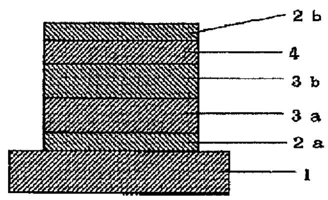
３ｃ　３ｂとは異なる化合物で構成される有機電子輸送層

４は界面層を表わす。

【図１】



【図２】



【図３】